An emissions-based view of climate forcing by methane and tropospheric ozone

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[1] We simulate atmospheric composition changes in response to increased methane and tropospheric ozone precursor emissions from the preindustrial to present-day in a coupled chemistry-aerosol-climate model. The global annual average composition response to all emission changes is within 10% of the sum of the responses to individual emissions types, a more policy-relevant quantity. This small non-linearity *between* emission types permits attribution of past global mean methane and ozone radiative forcings to specific emissions despite the well-known non-linear response to emissions of a single type. The emissions-based view indicates that methane emissions have contributed a forcing of $\sim$0.8–0.9 W m^{-2}, nearly double the abundance-based value, while the forcing from other ozone precursors has been quite small (~-0.1 for NO_x, $\sim$0.2 for CO + VOCs). **Citation:** Shindell, D. T., G. Faluvegi, N. Bell, and G. A. Schmidt (2005), An emissions-based view of climate forcing by methane and tropospheric ozone, *Geophys. Res. Lett.*, *32*, L04803, doi:10.1029/2004GL021900.

1. Introduction

[2] The climatically important reactive gases methane (CH_4) and ozone (O_3) are intricately coupled via atmospheric chemistry. Methane changes have both a direct radiative impact on climate and an indirect impact via their effect on ozone and OH. Similarly, emissions of the ozone precursors NO_x, CO and volatile organic compounds (VOCs, which here exclude methane) affect both tropospheric ozone and methane. Quantifying the contribution of emissions of each gas to the preindustrial (PI) to present-day (PD) composition changes, and hence radiative forcing, is challenging however, due to chemical non-linearities and the wide range of spatial and temporal scales involved in the chemical processes [*Kleinman*, 1994; *Prather*, 1994; *Klonecki and Levy*, 1997; *Grenfell et al.*, 2001]. Thus current assessments of climate change assume that forcing cannot be reliably and uniquely attributed to specific precursors, and instead rely on abundance-based calculations (i.e., based upon net final composition changes) [*Ramaswamy et al.*, 2001].

[3] We calculate radiative forcing due to anthropogenic emissions of methane and tropospheric ozone precursors using a fully coupled chemistry-aerosol-climate general circulation model (GCM) including methane chemistry and prescribed emissions. This work builds upon studies aimed at quantifying methane's indirect forcing via ozone [*Lelieveld et al.*, 1998; *Wang and Jacob*, 1998] and the net forcing from NO_x emissions [*Fuglestvedt et al.*, 1999; *Wild et al.*, 2001]. We demonstrate that non-linearities between emission types are quite small for the global annual average response to past emissions. Thus while the response to emissions of a single precursor may non-linearly depend on the initial composition state [*Stewart et al.*, 1977], the forcings from each precursor can be well separated from one another. The Global Warming Potential (GWP) of an emission pulse also includes both direct and includes indirect chemical effects [*Ramaswamy et al.*, 2001]. However, GWPs are not well-suited to large, sustained emissions changes such as the PI to PD changes, due to non-linearities in, for example, the O_3/NO_x relationship. Thus our estimates of the effects of past emissions are not derivable from either GWPs nor the observed abundance changes. Indeed, our emissions-based view of climate forcing gives a substantially different relative importance to various emissions than that suggested by current abundance-based assessments.

2. Model Description

[4] The Goddard Institute for Space Studies (GISS) tropospheric chemistry model [*Shindell et al.*, 2003] has been incorporated into the new, state-of-the-art GISS ModelE/Model III GCM. It includes the chemistry of HO_x-NO_x-O_x-CO-CH_4, hydrocarbon families and peroxyacetylnitrates. It contains complete sources and sinks for its 32 gases, including methane. The model is run at 4 by 5 degree resolution with 23 vertical layers, and the chemistry is fully interactive with the climate model and the GISS sulfate aerosol model (updated from *Koch et al.* [1999]). Soluble trace gases are intrinsically coupled to the GCM's hydrologic cycle. Full details and comparison with observations were presented for the previous version of the GCM [*Shindell et al.*, 2003]. The current chemical simulations are fairly similar, though some aspects are substantially improved in the more physically realistic tracer-climate model interaction of ModelE. For example, comparison of the annual cycle of ozone at five vertical levels (as in work by *Shindell et al.* [2003]) to a 16-site ozonesonde climatology [*Logan*, 1999] shows a reduction in the average absolute value monthly mean difference from 26% to 17%. For qualitative comparison, the average standard deviation of the observations is 34%. Differences have been reduced from 22% to 13% at 125 hPa, and from 22% to 15% at 200 hPa, indicating that ozone in the climatically critical tropopause region is quite well simulated. ModelE has eliminated the overly large wintertime downward ozone flux at high latitudes of the previous GCM while preserving the good agreement with ozone and precursor observations near the tropopause at lower latitudes [*Shindell et al.*, 2003]. The stratosphere-to-troposphere O_3 flux is 477 Tg/yr across

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Table 1. Response to Removal of Anthropogenic Emissions of the Indicated Gas[a]

	CH4 Response (ppbv)		O3 Response (Tg)		O3 Radiative Forcing (W m^{-2})
Perturbation	GISS GCM 100% Removal	GEOS-CHEM 50% Removal	GISS GCM 100% Removal	GEOS-CHEM 50% Removal	GISS GCM 100% Removal
CH4	−1209	−700	−51	−27	−0.20
NOx	422	167	−28	−21	−0.06
CO & VOCs[b]	−199	−72	−33	−8	−0.13
Sum of above	−986	−605	−112	−56	−0.39
All at once	−1084	−660	−99	−52	−0.37

[a]All values are global tropospheric (composition) or tropopause (forcing) annual mean change relative to the present-day.
[b]Results for GEOS-CHEM [*Fiore et al.*, 2002] are sums of separate studies reducing anthropogenic CO and VOCs.

150 hPa, in accord with the 450–590 Tg/yr range at 100 hPa estimated indirectly from satellite observations [*Gettelman et al.*, 1997]. Other gases are similar to the earlier model, with a good reproduction of the surface CO seasonal cycle, global-mean OH, and the CH_4 interhemispheric gradient but an exaggerated CH_4 seasonality at high northern latitudes.

[5] We performed simulations removing all anthropogenic emissions of CH_4, NO_x and CO + VOCs both individually and simultaneously. The reduction in methane emissions was 60%. All perturbations were calculated relative to PD to provide the most useful guide to the impact of potential reductions. Initial methane trends were extrapolated exponentially using the model's methane adjustment time (exponential decay time for a pulse) of 12.6 years. The steady-state turnover time for methane in our model is 9.4 years and so methane's recovery time is enhanced by 1.34. This enhancement, a diagnostic of tropospheric chemical stability, is in strikingly good agreement with the value of 1.35 obtained in the Met Office chemical transport model (CTM) [*Derwent et al.*, 2001], and in accord with theoretical predictions [*Prather*, 1994]. The extrapolated values were included following year 2 in 7 year runs, which in every case allowed methane (and ozone) to equilibrate. Results are averages over the last 3 years, compared with a PD control run. To permit standard calculations of radiative forcing without tropospheric response, methane and ozone changes were not allowed to affect the climate in these runs.

3. Linearity

[6] We examine linearity between precursors by comparing the methane and ozone responses to simultaneous changes in all precursors with the sum of the responses to individual changes. The responses in mass are within 15%, while radiative forcing varies by less than 10% (Table 1), indicating quasi-linearity in the chemical interactions between precursors. Though linearity was not addressed in their study, we find that a similar analysis of results from the independent GEOS-CHEM CTM [*Fiore et al.*, 2002] support our conclusions (Table 1). Both models simulated equilibrium conditions, however the short-term response may be non-linear.

[7] Additional evidence that non-linearities are small comes from studying the response to methane perturbations. Our model's response to the IPCC benchmark 10% increase in methane (∼175 ppbv) compares well with other estimates: tropospheric ozone increases by 0.58 DU and OH decreases by 2.9% compared with 0.40 to 0.85 DU for ozone and −2.7 to −3.3% for OH [*Prather et al.*, 2001]. We performed additional simulations using reductions in anthropogenic emissions of 10, 25, 50, 75, and 100 percent. The resulting Ozone and OH changes correlate linearly ($R > 0.99$) with emissions reductions, consistent with indications that the ozone yield per hydrocarbon oxidized has remained nearly constant since PI times [*Wang and Jacob*, 1998].

[8] Given this linearity, we can then examine interactions between precursor types. Simulation of the response to a 175 ppbv increase in methane abundance with PI conditions found an increase in tropospheric ozone of 0.69 DU and an OH decrease of 5.8%. Ozone's response to methane is proportional to the absolute methane change, while the OH response is proportional to the percentage change [*Prather et al.*, 2001]. The 175 ppbv increase represents a 25% increase over the PI value of 700 ppbv, so that the OH response normalized to a 10% increase relative to PI is then −2.3%, about 20% less than the PD value. The ozone response is ∼20% larger than for PD conditions. It thus appears that though both responses exhibit non-linearities going back to the PI atmosphere, the response did not varied dramatically. In contrast, ozone's sensitivity to NO_x changes alone may have been doubled in the past [*Wang and Jacob*, 1998].

[9] Thus while non-linearities certainly exist, particularly in the O_3/NO_x relationship and on shorter timescales and smaller spatial scales, we find that for interactions between precursor types they appear to have been minor for the PI to PD global average composition change. This is because pollutant levels at large scales have generally remained low enough that direct interactions between NO_x and hydrocarbons or CO (via HO_x), such as the reaction of NO_2 with OH, are not major loss pathways for odd nitrogen or hydrogen. This is especially true far from surface emissions, so that while the non-linearity in the ozone budget is 13%, the non-linearity in ozone's forcing, dominated by the region near the tropopause, is only 5% (Table 1).

4. Radiative Forcing

[10] The methane changes of 986–1084 ppbv are quite similar to the observed 1045 ppbv, while the ozone responses of 9.1–10.3 DU are consistent with other estimates of 6–13 DU [*Prather et al.*, 2001]. We have calculated the instantaneous tropopause radiative forcing due to emissions-induced changes in methane and ozone (Figure 1). The standard abundance-based view [*Ramaswamy et al.*, 2001] is shown for comparison. We use the standard calculation of radiative forcing from methane [*Ramaswamy et al.*, 2001]. For tropospheric ozone, the forcing is calculated within the GCM during the simulations, thus taking into account the spatial distribution of ozone changes. In contrast, CTM

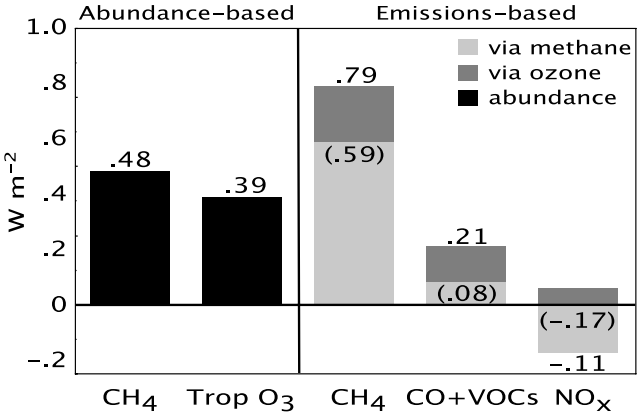

Figure 1. Radiative forcing from the preindustrial (1750) to the present-day (1998). Values above or below the bars give the total forcing, while values in parentheses give the forcing due to methane emissions alone. The abundance-based ozone value is from the sum of simulations with individual emission reductions for consistency with the emissions-based values. Forcing of ~0.1 W m^{-2} from stratospheric H_2O generated by methane is not included. For comparison, the abundance-based forcing from CO_2 is 1.46 W m^{-2}. Uncertainties in the abundance-based values are 0.13 for methane, based on the ~40% spread in forcing per unit methane estimates [*Hansen et al.*, 1997; *Ramaswamy et al.*, 2001], and 0.09 for ozone, based on the spread in model results [*Ramaswamy et al.*, 2001]. For emissions-based values, we estimate uncertainties by adding the direct methane forcing uncertainty given above in quadrature with the standard deviations in models' responses of ozone to individual emissions from *Prather et al.* [2001]. This yields 0.17 for methane, 0.10 for CO + VOCs (using the VOC value for CO), and 0.06 for NO_x. Uncertainty in the net forcing from non-methane ozone precursors is 0.11. Note that the emissions-based values are approximate, as the models of *Prather et al.* [2001] used substantially different perturbations than those used here, and the VOC perturbation was done simultaneously with CH_4 (we assume the uncertainty is dominated by VOCs).

studies have generally assumed a constant forcing per unit ozone change ranging from 34 to 48 mW m^{-2} DU^{-1} [*Hauglustaine and Brasseur*, 2001; *Wild et al.*, 2001; *Fiore et al.*, 2002]. In our simulations this ratio is 23 in response to NO_x emissions, and 43 for methane and CO + VOCs. This strong dependence upon type and location of emissions may account for the wide range of reported values, and suggests that this calculation should be done using a radiative transfer model (as in GCMs). The ozone response to NO_x emissions is largest from the surface to the mid-troposphere over Northern middle and high latitudes (Figure 2). Changes primarily occur close to the anthropogenic emissions in latitude and altitude since NO_x is short-lived and the extra-tropics are mostly NO_x-limited areas. In contrast, longer-lived CO and CH_4 cause ozone changes that maximize in the tropical upper troposphere (Figure 2), where present day ozone production is nearly NO_x saturated [*Jaeglé et al.*, 2001] and NO_x was relatively abundant even during the preindustrial. The radiative forcing maximizes for ozone near the tropopause and in the tropics, accounting for the weaker response per unit ozone from NO_x (note that when climate feedbacks are included, ozone at lower levels increases in importance [*Hansen et al.*, 1997]).

[11] Methane emissions have resulted in the largest forcing. The impact of methane emissions on ozone has caused a forcing of 0.2 W m^{-2}, while the impact on methane itself, which includes the chemical feedback of methane on its own lifetime, is 0.59 W m^{-2}. Even the latter value alone is 23% larger than the abundance-based value. This is because in the absence of other ozone precursor emissions, methane emissions alone would have lead to more methane than is currently present. Though the effect of other pollutants has offset some of this increase, the emissions viewpoint shows the true reduction in forcing that would be obtained from anthropogenic methane emissions reductions.

[12] Emissions of NO_x have had an overall negative forcing, as the induced reduction in methane outweighs the increased ozone, consistent with most earlier results [*Fuglestvedt et al.*, 1999; *Wild et al.*, 2001]. Our lower forcing per unit ozone change gives us a more negative response than the earlier studies, however. The response to CO and VOC emissions is an increase in both ozone and methane. The radiative impact from ozone is larger than from methane in this case, in accord with prior simulations of the response to increased emissions of industrial CO [*Wild et al.*, 2001].

5. Discussion

[13] In essence, we have calculated the direct plus indirect greenhouse response to emissions, as in GWPs, but for the specific case of the equilibrium response to reductions with the spatial distribution of our anthropogenic emissions. Were reductions to follow this path, our results and others [*Fuglestvedt et al.*, 1999; *Wild et al.*, 2001] suggest that NO_x decreases would increase global warming by inducing methane increases (though they are clearly desirable to improve air quality). Reductions in CO and VOC emissions would reduce both air pollution and climate forcing. However, the −0.79 W m^{-2} forcing from eliminating anthropogenic methane emissions would have the largest effect, 54% of the forcing from CO_2. This substantially exceeds the abundance-based methane forcing of 0.48 W m^{-2}. The total net forcing from non-methane ozone precursors is 0.10 W m^{-2}, only a quarter of the abundance-based tropospheric ozone forcing. Thus an emissions-based

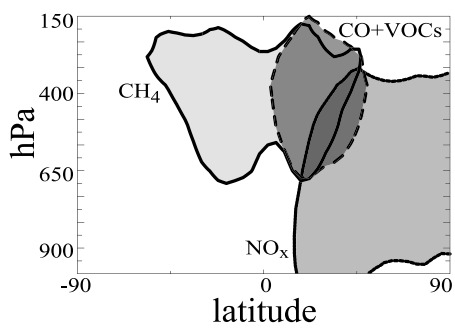

Figure 2. Ozone response to full removal of the indicated individual anthropogenic emissions. Contours show the area with 7 or more ppbv decrease in the zonal mean value.

view differs substantially from an abundance-based one, and by allowing attribution, adds to arguments for controls on methane emissions [*Hansen et al.*, 2000; *Fiore et al.*, 2002]. If the ~0.1 W m^{-2} forcing due to production of stratospheric H_2O from methane [*Hansen et al.*, 2000] were included, forcing from methane emission would be 61% of the forcing from CO_2.

[14] Though non-linearities between precursors appear to have had a minor role in past global-average atmospheric evolution, this will not necessarily be the case in the future, and forcing reductions from any particular control strategy should be calculated with a full model. Additionally, impacts of reductions will likely vary greatly from region to region [*Klonecki and Levy*, 1997; *Fuglestvedt et al.*, 1999; *Derwent et al.*, 2001] though less so for long-lived methane. For the past, however, the global average forcing uncertainties resulting from non-linear chemical coupling appear to be around 10%, much smaller than the 52% spread in abundance-based model-derived tropospheric ozone forcing estimates or the 40% spread in forcing per unit methane estimates [*Jain et al.*, 2000; *Ramaswamy et al.*, 2001]. Changes other than precursor emissions such as a mildly warmer climate or increased sulfate aerosols have likely had a minimal impact on chemical changes [*Grenfell et al.*, 2001]. This suggests that an emissions-based view provides useful insight for policy-making, in conjunction with GWPs, while introducing little additional uncertainty in comparison with the standard abundance-based view.

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